

# Assessment of the Transit Time of Sulfate in Forest Soils Using Isotope Techniques

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The transit time of sulfate in forested ecosystems is a matter of considerable debate and previous estimates have ranged from conservative behavior<sup>1</sup> to retention times of many decades<sup>2</sup>. A potential tool for tracing sulfur (S) fluxes in the environment is the radioactive isotope <sup>35</sup>S. However, because of its short half live time of 87 days, this isotope provides only meaningful results for relatively short S transit times of less than two years<sup>3</sup>. Applications of <sup>34</sup>S enriched or depleted sulfate compounds constitute an alternative approach for assessing the transit time of sulfate in the environment<sup>4</sup>. In contrast to the radioactive isotope <sup>35</sup>S, the tracer technique based on stable isotopes has the advantage that it is applicable to long term studies, if the tracer has distinct <sup>34</sup>S/<sup>32</sup>S ratios and is applied in sufficient amounts.

In July 1990, an equivalent of 70 kg S ha<sup>-1</sup> was applied as K<sub>2</sub>SO<sub>4</sub> solution to a forest floor (Orthic Luvisol) near Munich, Germany. The SO<sub>4</sub><sup>2-</sup> was derived from Silurian gypsum with a δ<sup>34</sup>S value of +26 ‰, whereas S in soil and seepage water before tracer application had δ<sup>34</sup>S values near 0 ‰. Since isotope discrimination is believed to be small during S transformation processes under aerated conditions<sup>5</sup>, the large difference between the δ<sup>34</sup>S values of the K<sub>2</sub>SO<sub>4</sub> and the ecosystem S was considered to be ideal for tracing the fluxes and transformations of the labeled sulfate. The objective of the study was to determine the transit time of sulfate in the uppermost meter of the forest soil by observing the movement of the isotopically labeled sulfate through the unsaturated soil zone.

Throughout the first 2.5 years after tracer application, seepage water was sampled underneath the forest floor (5 cm) and in the mineral soil at 20 and 100 cm depths. Sulfur isotope ratios for seepage water sulfate collected at 5 cm depth indicated that the tracer passed through the forest floor within four months after application. Mass and isotope balances showed that approximately 50 kg tracer S, or approximately 70% of the applied K<sub>2</sub>SO<sub>4</sub>, had passed the mineral soil horizons in 20 cm depth 2.5 years after tracer application. No labeled sulfate was detected in 100 cm soil depth during the observation period indicating that the bulk of the tracer S must have been retained in the lower mineral soil horizons of the Orthic Luvisol.

Isotope ratio determinations on various soil sulfur compounds were performed on samples obtained from various depths in May 1990 (prior to tracer application), in 1991, 1995, and 1999. Results show that little tracer S was retained in the humus layers and Ah horizons, which contain S dominantly in organic binding form. In 1991, tracer S was detected between 5 and 40 cm soil depth predominantly as inorganic sulfate. In 1995, the bulk of the tracer S was detected between 15 and 70 cm soil depth mainly as inorganic sulfate. Between 1995 and 1999, little tracer movement was evident from the isotope ratios of total soil S and it appeared that the labeled S had not reached 100 cm soil depth nine years after tracer application.

Preliminary data from this ongoing study provide evidence that the mean residence time of sulfate in the uppermost meter of the investigated Orthic Luvisol clearly exceeds one decade.

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